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A study of particulate emissions during 23 major industrial fires: Implications for human health

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ABSTRACT

Public exposure to significantly elevated levels of particulate matter (PM) as a result of major fires at industrial sites is a worldwide problem. Our paper describes how the United Kingdom developed its Air Quality in Major Incidents (AQinMI) service to provide fire emission plume concentration data for use by managers at the time of the incident and to allow an informed public health response. It is one of the first civilian services of its type anywhere in the world. Based on the involvement of several of the authors in the AQinMI service, we describe the service's function, detail the nature of fires covered by the service, and report for the first time on the concentration ranges of PM to which populations may be exposed in major incident fires. We also consider the human health impacts of short-term exposure to significantly elevated PM concentrations and reflect on the appropriateness of current short-term guideline values in providing public health advice. We have analysed monitoring data for airborne PM ($\leq 10 \mu\text{m}$, PM_{10} ; $\leq 2.5 \mu\text{m}$, $\text{PM}_{2.5}$ and $\leq 1.0 \mu\text{m}$, PM_1) collected by AQinMI teams using an Osiris laser light scattering monitor, the UK Environment Agency's 'indicative standard' equipment, during deployment to 23 major incident industrial fires. In this context, 'indicative' is applied to monitoring equipment that provides confirmation of the presence of particulates and indicates a measured mass concentration value. Incident-averaged concentrations ranged from 38 to $1450 \mu\text{g m}^{-3}$ for PM_{10} and 7 to $258 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$. Of concern was that, for several incidents, 15-min averaged concentrations reached $> 6500 \mu\text{g m}^{-3}$ for PM_{10} and $650 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$, though such excursions tended to be of relatively short duration. In the absence of accepted very short-term (15-min to 1-h) guideline values for PM_{10} and $\text{PM}_{2.5}$, we have analysed the relationship between the 1-h and 24-h threshold values and whether the former can be used as a predictor of longer-term exposure. Based on this analysis, for PM_{10} , our tentative 1-h threshold value for use in deciding whether to close public buildings or to evacuate areas is $510 \mu\text{g m}^{-3}$. For $\text{PM}_{2.5}$, 1-h concentrations exceeding $350 \mu\text{g m}^{-3}$ might indicate longer-term exposure problems. We conclude that whilst services such as AQinMI are a positive development, there is a need to consider further the accuracy of the data provided and for the development of very short-term guideline values (i.e. minutes to hours) that responders can use to determine the appropriate public health response.

1. Introduction

Episodic, acute, exposure of populations to airborne particulate matter (PM, where: $\leq 10 \mu\text{m}$, PM_{10} ; $\leq 2.5 \mu\text{m}$, $\text{PM}_{2.5}$ and $\leq 1.0 \mu\text{m}$, PM_1) at concentrations in the hundreds, and even thousands, of micrograms per cubic metre can occur under a variety of different scenarios, including: forest fires (Delfino et al., 2009; Heil and Goldammer, 2001; Sastry, 2002), dust storms (Godri et al., 2011; Hefflin et al., 1994; Karanasiou et al., 2012; Lee et al., 2013; Pey et al., 2013; Sajani et al.,

2011; Stafoggia et al., 2016; Vodonos et al., 2014), crop residue burning (Gupta et al., 2016; Yang et al., 2008), festivals and celebrations involving fireworks (Barman et al., 2008; Beig et al., 2013; Godri et al., 2010; Wang et al., 2007), volcanic eruptions (Horwell and Baxter, 2006; Nania and Bruya, 1982), and industrial/urban emissions under adverse meteorological conditions (Macintyre et al., 2016; Schwartz, 1994; Zhou et al., 2015). These episodes, as summarised in Table 1, are known to have adverse health impacts, with probably the most well-known example being the London smog episode of 1952 when total

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Table 1
Average daily particulate concentrations, and associated health impacts, recorded during air pollution episodes arising from various sources.

No.	Source and location	Year(s)	Maximum average daily particulate concentration ($\mu\text{g m}^{-3}$)	Health impacts Note: RR = Relative Risk	Ref
1	Atmospheric particulate matter episode, London	1952	2000 (TSP) ^a	RR = 1.06 for all-cause mortality per $100 \mu\text{g m}^{-3}$ increase	Schwartz (1994)
2	Atmospheric particulate matter episode, Ji County in Tianjin, China	2013	607 ($\text{PM}_{2.5}$)	RR = 1.069 (Confidence interval, CI 0.998 to 1.150) for all-cause mortality following a smog episode; RR = 1.013 (CI 1.0074 to 1.019) for all-cause mortality per $10 \mu\text{g m}^{-3}$ increase in $\text{PM}_{2.5}$.	Zhou et al. (2015)
3	Atmospheric particulate matter episodes, United Kingdom	2014	83 ($\text{PM}_{2.5}$)	Two episodes (10 days total) associated with 600 deaths and 1570 emergency respiratory and cardiovascular admissions.	Macintyre et al. (2016)
4	Forest Fires (S.E. Asia), measured in Kuala Lumpur, Malaysia.	1997	424 (PM_{10})	RR = 1.19 for all-cause mortality the day after a $> 210 \mu\text{g m}^{-3}$ event	Sastry (2002)
5	Californian wildfires, measured at Forest Fires (S.E. Asia), measured in Palangkaraya, Indonesia.	2003 1997	76 ($\text{PM}_{2.5}$) 4000 (TSP)	RR = 1.143 for respiratory admissions per $10 \mu\text{g m}^{-3}$ See item 4	Delfino et al. (2009) Heil and Goldammer (2001)
7	Forest Fires (S.E. Asia), measured in Kuching, NW Borneo-Malaysia.	1997	930 (PM_{10})	See item 4	Heil and Goldammer, (2001)
8	Seasonal dust storm, Southeast Washington State	1991	> 1000 (PM_{10})	3.5% and 4.5% increases in emergency room visits for bronchitis and sinusitis respectively per $100 \mu\text{g m}^{-3}$ increase in PM_{10} .	Hefflin et al. (1994)
9	African dust outbreaks, measured in the Emilia-Romagna region of Italy	2002 to 2006	100 (PM_{10})	Respiratory mortality increased by 22% on dust storm days for people over 75.	Sajani et al. (2011)
10	African dust outbreaks, measured across the Mediterranean basin	2004	300 (PM_{10})	Not determined	Pey et al. (2013)
11	African dust outbreaks, measured in Be'er Sheva, Israel	2001 to 2010	4797 (PM_{10})	RR = 1.16 for hospitalisation due to chronic obstructive pulmonary disease (COPD) exacerbation on dust storm days ($> 71 \mu\text{g m}^{-3}$)	Vodonos et al. (2014)
12	African dust outbreaks, measured in Les Palmas, Gran Canaria.	2001 to 2005	612 (PM_{10}); 242 ($\text{PM}_{2.5}$)	RR = 1.22 (CI 1.07 to 1.39) for asthma admissions per $10 \mu\text{g m}^{-3}$ increase in $\text{PM}_{2.5}$.	Lopez-Villarrubia et al. (2016)
13	Fireworks, Beijing, China	2006	466 (PM_{10}); 184.3 ($\text{PM}_{2.5}$) (12 h sampling period)	Not determined	Wang et al. (2007)
14	Fireworks, Lucknow, India	2005	963 (PM_{10})	Not determined	Barman et al. (2008)
15	Fireworks, London	2007	60 (PM_{10})	Not determined	Godri et al. (2010)
16	Crop residue burning, Suqian, China	2006	340 (PM_{10})	Not determined	Yang et al. (2008)
17	Crop residue burning	2013 to 2014	167 (PM_{10}) 107 ($\text{PM}_{2.5}$)	Decrease of between 4 and 6% in Peak Expiratory Flow during burning periods.	Gupta et al. (2016)
18	Volcanic eruption, Mount St Helens	1980	3000 to 33,000 (TSP)	10% increase in hospital visits	Horwell and Baxter (2006), Nania and Bruya (1982)

^a TSP can be converted to PM_{10} using a factor of 0.55 (Pope et al., 1995).

suspended particulate (TSP) concentrations reached $2000 \mu\text{g m}^{-3}$ over a five-day period (Pope et al., 1995; Schwartz, 1994). Over the duration of the episode, there were an estimated 4000 deaths attributed to the high pollution levels (Logan, 1953), with these coming almost exclusively from cardiovascular and respiratory disease (Bell and Davis, 2001). There were an estimated additional 7300 deaths over the subsequent months (Bell and Davis, 2001). Nevertheless, there is evidence from the episodes listed in Table 1 that not all acute exposures produce similar health effects: natural dust events (entries 8 to 12) or volcanic eruptions (entry 18) appear to be associated mainly with increased hospital visits rather than mortality. Schwartz et al. (1999), for example, analysed the health effects of 17 dust storms in Spokane, Washington (average 24-h mean $\text{PM}_{2.5}$ concentration of $263 \mu\text{g m}^{-3}$) and found no evidence for increased mortality. One explanation for the differences in health impacts observed for natural and combustion-related PM is that the latter has a higher $\text{PM}_{2.5}$ fraction, with may contain elevated soluble transition metal and organic compound content; these are thought to be important contributing factors in proposed oxidative stress mechanisms for ill health caused by PM exposure (Kelly and Fussell, 2012). Such conclusions are supported by short-term ambient studies from across the developing and developed world, where traffic is the main source of fine PM. These studies have consistently shown that a $10 \mu\text{g m}^{-3}$ increase in PM_{10} concentration produces a 0.5% increase in short-term mortality (World Health Organisation, 2006), similar to the mortality rates observed for the combustion-related sources in Table 1. Based on these associations, the World Health Organization (WHO) have set short-term (24-h mean) ambient air quality guideline values for PM_{10} and $\text{PM}_{2.5}$ of $50 \mu\text{g m}^{-3}$ and $25 \mu\text{g m}^{-3}$ respectively. There is no standard set for PM_{10} .

1.1. Major incident fires

An additional, but little studied, source of significantly elevated PM concentrations arises from uncontrolled open fires involving industrial or commercial premises which, if located near to residential areas, will represent a clear risk to public health from exposure to the plume (Lemieux, 2002; Stec, 2017). In the current work, we discuss the approach of the United Kingdom (UK) in dealing with such incidents, through their 'Air Quality in Major Incidents' (AQinMI) service. The service has operated from 2009 to the present date with the purpose of responding to 'major incident' fires, providing a coordinating role and facilitating the collection and dissemination of representative modelled or monitored airborne contamination data to support a public health risk assessment. Three of the authors have been involved in this service, including overall incident management, and management of one of the contracted monitoring teams (between April 2009 and March 2014 Northumbria University coordinated the AQinMI monitoring team for the North of England on behalf of the UK Environment Agency). In the remainder of this section, we describe the evolution of this service.

The magnitude of the public health risk from any uncontrolled major incident fire depends on the scale of the fire, composition of its fuel source, duration and temperature of burn, ventilation conditions, firefighting methods used, proximity to human populations, and plume dispersal characteristics, as influenced by the prevailing meteorological conditions (Environment Agency, 2009; Powlesland, 2008; World Health Organisation, 2009). Depending upon the source of the fire, the fire emission plume will comprise a complex mixture of gaseous airborne toxicants, including: (a) asphyxiants, such as carbon monoxide (CO), hydrogen cyanide (HCN), and carbon dioxide (CO_2), (b) irritants, including sulfur dioxide (SO_2), nitrogen oxides (NO_x), phosphorus pentoxide (P_2O_5), hydrogen chloride (HCl), hydrogen fluoride (HF), hydrogen bromide (HBr), acrolein and formaldehyde, and (c) a range of complex organic contaminants including polycyclic aromatic hydrocarbons (PAHs), dioxins and dibenzofurans (Purser et al., 2016; Wakefield, 2010). PM is also produced, and these may contain adsorbed metals and organic contaminants. Additionally, specific toxic

substances may be released from ruptured tanks and drums that have not undergone combustion (Wakefield, 2010). As well as the toxicological health risks posed by airborne pollutants released from 'major incident' fires, there will be a direct risk to health from fire and explosions. Members of the affected population may also experience anxiety and other stress disorders due to the proximity of an often very visible fire or as a result of having to be relocated away from their homes (Baxter et al., 1995). Consequently, there is a clear requirement for national and local public health services to provide the necessary infrastructure and organisation to manage major fire incidents in a coordinated and resilient manner. Such a service should be able to determine the composition and toxic effects of a plume, give timely advice on the risk of exposure to populations and emergency service personnel and calculate plume behaviour under various weather conditions (Baxter et al., 1995). This remit is consistent with World Health Organization (WHO) advice that advocates a role for public health practitioners in risk assessment and communication during emergency situations. The WHO identifies a four-step procedure for such a role: (1) hazard identification, (2) establishing dose-response relationships, (3) carrying out exposure assessments and (4) risk characterisation (Stewart-Evans et al., 2014; World Health Organisation, 2009).

In the UK, prior to 2009, an appropriate coordinated civilian 'major air pollution incident' response service did not exist as demonstrated by the public health response to the 2005 explosion and fire at an oil storage and transfer depot in the Buncefield, Hertfordshire (World Health Organisation, 2009). The Buncefield depot burned for five days, leaving most of the site destroyed and releasing a plume of combustion products to the atmosphere that dispersed across southern England and beyond to mainland Europe (Buncefield Major Incident Investigation Board., 2008; Vautard et al., 2007). To evaluate the UK's response to this incident, and the lessons to be learned, a Major Incident Investigation Board (MIIB) was established (Buncefield Major Incident Investigation Board., 2008). The principal shortcomings identified by the investigation drove a change in major air pollution incident management in the UK and introduced the concept of the 'Air Quality Cell' (AQC), a mechanism, reflective of WHO Guidance, to bring together various state agencies to deliver public health risk assessment advice to support community resilience (World Health Organisation, 2009). In doing so, it built upon the existing public health infrastructure, particularly the UK Health Protection Agency (HPA, now Public Health England) and its specialist Chemical Hazards and Poisons Division (now Environmental Hazards and Emergencies Department) (Palmer and Coleman, 2013). Whilst these bodies were already established with the purpose of providing health protection advice, they lacked the ability to model and monitor the plume arising from Buncefield (Buncefield Major Incident Investigation Board., 2008; Palmer and Coleman, 2013). Consequently, appropriate and timely advice for incident managers about the health impact of exposure to the products of combustion, and the implications of firefighting techniques were not available for the Buncefield incident. The investigation also proposed a review of the mechanism for 'obtaining and using air quality data in an emergency' (Buncefield Major Incident Investigation Board, 2007). This review was to focus on agreeing: (1) notification and cascade procedures; (2) agency roles and responsibilities for collecting air quality data; (3) arrangements to disseminate air quality data to responders; and (4) how to include them in emergency planning and response.

1.2. Establishing a public health response mechanism for major incidents fires

Commencing on a trial basis in April 2009, and more formally from April 2010, the Environment Agency (EA) in England and Wales, and the Scottish Environment Protection Agency (SEPA) in Scotland became responsible for coordinating the sourcing of modelled and monitored air quality data during 'major incident' fires as part of the AQinMI Service (McParland and Paranthamanm, 2012), though fires involving

the release of chemical, biological, radiation or nuclear (CBRN) warfare agents were purposefully excluded from this service. Monitoring instruments deployed in these events offer an ‘indicative standard’ rather than a reference one. The term ‘major incident’ is a label reserved for situations that pose a serious threat to public health, or cause a significant number or type of casualties, such that special arrangements are needed to manage them (NHS England, 2015). In the specific context of fires, ‘major incidents’ are those that are likely to have a duration of > 6 h (Barker, 2010; Powlesland, 2008) and for which there is a risk to public health and, by implication, the environment (note that the 6 h threshold is only intended to be used as a guideline by the EA national duty officer to determine the worthwhileness of activating monitoring teams when it might take several hours for them to be mobilised, travel to site, and to set up and obtain meaningful data). The overall aim of the AQinMI service is to characterize the plume and indicate its impact on human health and well-being (Lawrence, 2015) so as to determine whether exposure levels are safe for people to go about their daily activities in areas where precautions are not being taken, or to confirm that areas are safe following periods where sheltering or evacuation had been advised. Provided the AQC is capable of monitoring the substances emitted, the AQC’s responsibility can extend beyond plumes from major fire incidents to include loss of containment incidents such as at chemical works.

AQCs comprise the public bodies identified in Table 2 (Tyne and Wear Fire and Rescue Authority, 2010). The actual monitoring is undertaken by private contractors, engaged by the Environment Agency, who are each equipped with the agreed AQinMI service monitoring equipment so that each monitored incident is provided with a consistent source of air pollution data during the acute incident (detailed in Table 3).

The initial phase of the AQinMI service ran between April 2009 and March 2014 with England and Wales being divided into eight regions with seven teams responsible for field monitoring of air pollutants and for relaying that data back to the AQC. This data is used by Public Health England (PHE) to provide an opinion as to the potential threat to public health and fed into the management of the response. The seven field monitoring teams were capable – critically for such time-sensitive data – of being rapidly mobilised on a 24/7 basis. Northumbria University was one such contractor, with responsibility for the North of England. In addition, two mobile laboratories, which were slower to deploy and set-up but contained reference standard monitoring equipment, were available.

1.3. Operation of the AQinMI service

There are three levels of activation of the AQinMI service. Firstly,

there are PHE, EA and Met Office national duty staff, who are available 24/7 to support local responders (with PHE and the EA national duty staff each advising their own local staff, and the Met Office providing meteorological forecasts to those who need them). The local staff are the ones initially dealing with the incident in liaison with emergency services, local authorities and other response partners. Based on this local engagement and insight, the PHE and EA national duty officers keep under constant review the need to escalate the service to an AQC and whether or not monitoring and sampling might be required. The AQinMI service is, therefore, engaged on a regular basis to support local staff and responders with specialist advice and background risk assessment.

The second level of activation is escalation to an AQC. Where significant health or environmental risks might be presented by emissions from an incident, the HE and EA national duty officers will jointly agree to convene an AQC to provide an expert forum for the ongoing assessment of those risks and to give advice to those managing the incident on the ground (via their convened multi-agency co-ordination groups). The core members of an AQC are on 24/7 standby, and it can be convened within 1 to 2 h of being alerted to a relevant incident.

The final level of activation occurs when AQC members decide they need monitoring data to inform their ongoing risk assessments and to validate decision making and public health advice. Monitoring locations are agreed between the EA and PHE (Environmental Hazards and Emergencies Department) in advance of the contractor arriving at an incident. Decisions are based on: (a) modelled plume behaviour using the Meteorological Office’s chemical meteorological forecasts (known as CHEMETs, generated using the Numerical Atmospheric Dispersal Modelling Environment (NAME III) modelling software (Leadbetter et al., 2010; Meteorological Office, 2017); (b) the availability of appropriate services e.g. electrical supply for monitoring equipment (Izon-Cooper, 2010), and (c) occupational health and safety concerns for the monitoring team themselves. Deployment locations may move during the course of an incident due to changes in local circumstances, e.g. wind direction, etc. but the number of times this happens must be limited because of the time taken to dismantle instruments, move them and set them up again.

A full list of monitoring/sampling equipment carried by the response teams and the range of determinands that can be tested for is given in Table 3. In practice, the Gasmeter and Osiris instruments are setup first and left running for the duration of the incident (data was downloaded and sent to the EA every 2 h), with other continuous methods and sampling techniques deployed as necessary. Samples collected by filter and impinger solutions during the incident are sent for laboratory analysis, and the results are used to inform future incidents of this type. Continuous monitoring data is the main

Table 2

Details the multiple agencies that contribute to the development of public health advice during an AQC, and their roles for the initial period of the service (April 2009 to March 2014).

Agency	Role in an AQC
Environment Agency	<ul style="list-style-type: none"> Overall responsibility for the management of AQCs. Responsible for the provision of monitoring data using contracted rapid response teams across England and Wales Specification of hand-held monitoring and sampling instruments Maintenance of two monitoring and sampling response vehicles Access and download other data from national air quality networks Chair individual AQCs
Public Health England Meteorological Office	<ul style="list-style-type: none"> Provides public health advice based on modelled and monitoring data. Provides weather predictions. Provides modelled air quality information.
Food Standards Agency Health and Safety Laboratory Local Authorities	<ul style="list-style-type: none"> Strategic responsibility for contamination of foodstuffs, e.g. crops Ability to run more complex source term and plume dispersion models for more complex or prolonged incidents. No duty to engage in an AQC but can be invited to do so. Extent of engagement may be dependent on the priority for air pollution management under the UK’s National Air Quality Strategy, and so they may have local air quality monitoring data to support the public health risk assessment Have local knowledge including that about local industry, or sensitive receptors. Take on responsibility to manage the response once the ‘recovery’ phase commences.
Independent laboratory contractor	<ul style="list-style-type: none"> Laboratory analysis of samples collected from impingers, etc.

Table 3

Details of monitoring equipment used in 'major incident' fires.

Monitoring equipment	Principle	Determinands
Continuous monitoring		
Turnkey Osiris particulate monitor	Laser light scattering (670 nm)	TSP, PM ₁₀ , PM _{2.5} , and PM ₁
Gasmeter DX4030	Infrared	water, carbon dioxide, carbon monoxide, nitrous oxide, methane, sulfur dioxide, ammonia, hydrogen chloride, hydrogen bromide, hydrogen fluoride, hydrogen cyanide, formaldehyde, 1,3-butadiene, benzene, toluene, ethyl benzene, m-xylene, o-xylene, p-xylene, acrolein, phosgene, arsine, phosphine and methyl isocyanate.
QRAE	Electrochemical cell	Chlorine and carbon monoxide.
Arizona Instruments Jerome 631 analyzer	Gold film analyzer (electrical resistance)	Hydrogen sulfide.
Casella NOMAD portable weather station	–	Temperature, relative humidity, pressure, wind speed and direction, solar radiation and rainfall.
Monitoring with sample pumps/media		
Tecora Delta low flow pump	Impinger (0.05 M sodium hydroxide solution) Impinger (0.05 M sulfuric acid solution) PTFE + silver membrane Silica gel – Supelco Orbo 53 Thermal desorption (TD) tube	Hydrogen cyanide, acetic acid, hydrogen sulfide, chromic acid. Ammonia. Bromine and chlorine. Hydrogen fluoride, nitric acid, phosphoric acid, sulphuric acid, sulfur trioxide and arsine. 1,1,1-trichloroethane, 1,2-dichloroethane, 1,3-butadiene, 2,4-toluene diisocyanate, 2,6-toluene diisocyanate, acetone, acetonitrile, acrolein, acrylamide, acrylonitrile, benzene, carbon disulfide, chlorobenzene, chloroform, chloropicrin, dichloromethane, ethyl acrylate, ethyl benzene, ethyl isocyanate, ethylene oxide, formaldehyde, methyl acrylate, methyl bromide, methyl chloride, 2-butanone, methyl isocyanate, methyl isothiocyanate, methyl methacrylate, methyl styrene, phenol, phosgene, propane, styrene, tetrachloroethylene, tetrachloromethane, toluene, trichloroethylene, vinyl chloride, xylene, other volatile organic compounds.
Tecora Echo high volume sampler	Asbestos filter Quartz filter PUF plug	Asbestos Antimony, arsenic, cadmium, chromium, lead, manganese, nickel, platinum, thallium, vanadium, mercury, other metals, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, pesticides. Dioxins and derivatives, including polychlorinated dibenzodioxins, furan and derivatives, including polychlorinated dibenzofurans.

information from which public health protection risks are determined. The UK's devolved jurisdictions of Northern Ireland, Scotland and Wales now operate their own variants of the AQC.

In this paper, we report the magnitude of PM emissions from 23 major incident AQC fires covering the period from April 2009 to March 2016, involving a range of commercial or industrial facilities, and identify the need for short-term exposure metrics. Whilst responses to a limited number of individual incidents have previously been reported in Public Health England's Chemical Hazards and Poisons Reports, there is no other literature that has outlined the function and evolution of the AQinMI service, or provides an analysis of the Osiris PM monitoring data derived from the range of AQC major air pollution incidents.

2. Methodology

2.1. Source of AQC particulate monitoring data

Osiris monitoring data for PM₁₀, PM_{2.5}, and PM₁ from the AQC events reported in this paper was sourced from the UK's Environment Agency for the period April 2009 to March 2016; this included monitoring data collected by Northumbria University's North of England AQinMI monitoring team.

The method used by AQinMI teams to monitor the time series of PM₁₀, PM_{2.5} and PM₁ concentrations during each of the 23 major incident fires was the Osiris PM monitor (a non-reference standard monitor, certified to the UK EA MCERTS performance standard for indicative PM monitors, for PM₁₀). The Osiris monitor utilises a light scattering technique to identify the mass concentration of TSP, PM₁₀, PM_{2.5} and PM₁ within the range of 0.4 to 20 µm with a resolution of 0.01 µg m⁻³. Air is drawn through a heated inlet into the Osiris monitor with a flow rate of 0.6 L minute⁻¹ allowing only one particle to be illuminated by the laser light beam (670 nm) at any moment. > 20,000 particles per second can be sized before coincidence occurs.

Typically, this corresponds to a concentration > 6000 µg m⁻³. The effective operating temperature is – 5° to + 40 °C. Light diffraction for individual particles is converted to an electrical pulse proportional to the size of the particle giving an equivalent mass for a given period, between 1 s and 4 h, using a look-up table. The Osiris only measures diffraction angles between 0 and 10°, within which range diffraction is independent of PM composition (Turnkey Instruments, 2009; Deary et al., 2016).

Monitoring locations used during these incidents reflected the location of sensitive receptors, for example, schools, hospitals or residential areas and was decided upon by the AQC. Short breaks in monitoring did occur periodically due to data download/sharing with the EA and also due to the relocation of monitoring crews, for example in response to changes in wind direction.

2.2. Data analysis

15-min averaged values were calculated from the original AQC measurement data, which comprised one-minute averages. The justification for averaging the data over 15 min is that: (a) this sampling period has been applied to measure the within-day variability of PM concentrations (Godri et al., 2011; Ramachandran et al., 2003), and (b) this time period also corresponds to the usual resolution of tapered element oscillating microbalance (TEOM) and TEOM-FDMS (Filter Dynamics Measurement System) instruments (Godri et al., 2011). Further, there is evidence that short-term particle excursions, undetected when using 24-h averaging periods that correspond to WHO and EU standards, may have significant health effects, particularly for individuals with underlying medical conditions such as asthma (Michaels and Kleinman, 2000). Moreover, evidence from healthy human volunteers exposed to diesel exhaust fumes has demonstrated a measurable lung inflammatory response to short-term (2-h) exposure (Behndig et al., 2006). The 15-min averages were used to produce box and

Table 4
List of all AOCs managed by the UK Environment Agency. It reflects all AOCs declared in England for the period April 2009 to March 2016 and in Wales between April 2009 to March 2016.

AQC No. ^a	Date	Location ^b	Site operation ^c	Monitoring data available
1	13 May 2009	Riverside Recycling Waste Limited, Hull (NGR: TA 10069 31,417)	Waste accumulation	Yes
2	17 June 2009	Skymark Packaging International Limited, Southern Avenue, Leominster, Herefordshire HR6 0QF (NGR: SO 50035 58,135)	Packaging manufacturer	No
3	21 August 2009	Murfit Industries, Wisbech Road, Littleport, Ely, Cambridgeshire CB6 1RA (NGR: TL 54808 87,704)	Tire recycling facility	Yes
4	08 September 2009	Kingpin Tyres Limited, Wem Industrial Estate, Souton Rd., Wem, Shrewsbury SY4 5SD (NGR: SJ 52485 29,917)	Tire recycling facility	Yes
5	02 October 2009	Wincanton WEEE Recycling, Fawsley Drive, Heartlands Business Park, Daventry, Northamptonshire NN11 8UG (NGR: SP 56016 64,359)	WEEE recycling facility	No
6	05 January 2010	Countrystyle Group Parham Recycling Centre, Parham, Woodbridge, Suffolk, IP13 9AF (NGR: TM 32956 61,030)	Recycling and composting facility	Yes
7	21 January 2010	Farm, Higher Burghill Farm, Wotter, Devon PL17 5HU (NGR: SX 54770 62,233)	Barn fire involving chemical fertilizer	No
8	21 April 2010	Interfloor Limited, Littleburn Industrial Estate, Langley Moor, County Durham, DH7 8HJ (NGR: NZ 25475 39,913)	Tire recycling facility	Yes
9	24 May 2010	Grosvenor Chemicals, Lees Mill Lane, Linthwaite, Huddersfield, West Huddersfield HD7 5QE (NGR: SE 09005 14,185)	COMAH site, chemical processing	Yes
10	16 June 2010	Solvent Safety, Plumtree Farm Industrial Estate, Plumtree Rd., Bircotes, Doncaster DN11 8EW (NGR: SK 63265 92,532)	Hazardous waste transfer station	No
11	24 June 2010	Viridor Waste Management, Horningtops, Liskeard PL14 3QD (NGR: SX 26361 60,855)	Landfill taking other wastes	No
12	28 June 2010	Illegal waste dump, Units 1–4 Whitelea Grove Lee Industrial Park, Mexborough, Doncaster, South Yorkshire S64 9QP (NGR: SK 46626 99,883)	Waste accumulation	Yes
13	05 July 2010	Kingpin Tyres Limited, Wem Industrial Estate, Souton Rd., Wem, Shrewsbury SY4 5SD (NGR: SJ 52485 29,917)	Tire recycling facility	Yes
14	12 July 2010	Claddagh Recycling Limited, Neath Abbey Wharf, Skewen, Neath, West Glamorgan, SA10 6BL (NGR: SS 72780 96,029)	Waste management facility	Yes
15	11 August 2010	BCB Environmental (UK) Limited, The Green Hangar, Unit 87 Marston Business Park, Tockwith, YO26 7QF (NGR: SE 45597 52,363)	Hazardous waste management: transfer station	No
16	02 May 2011	Sunline Direct Limited, Cotton Way, Loughborough, Leicestershire LE11 5FJ (NGR: SK 51974 21,120)	Direct mailing company	No
17	16 June 2011	Fforestfach (illegal stockpile), Unit 1, Queensway, Fforestfach Industrial Estate, Swansea (NGR: SS 62778 95,721)	Tire waste dump	Yes
18	11 October 2011	THINK Environmental, Blackbridge Farm, Cranford Road, Kettering, NN15 5JJ (NGR: SP 90942 76,374)	Waste management facility	No
19	12 August 2012	Hunts Waste Recycling, 75–77 Chequers Lane, Dagenham, RM9 6QJ (NGR: TQ 48851 82,189)	Waste management facility	Yes
20	14 January 2013	Lewis Skip Hire, Milfraen View, Blaenau View, Nantyglo, Ebbw Vale, Gwent NP23 4PQ (NGR: SO 19308 10,919)	Waste management facility	Yes
21	16 June 2013	Lawrence's Waste Transfer Station, Stourport Road, Kidderminster, Worcestershire DY11 7QF (NGR: SO 81974 73,720)	Waste management facility	Yes
22	30 June 2013	J & A Young (Leicester) Limited, Dartmouth Road, Smethwick, Birmingham, B66 1AS (NGR: SP 02415 88,629)	Waste management facility: plastics recycling	Yes
23	21 August 2013	J25 Recycling Limited, Oldmoor Road, Bredbury, Stockport (NGR: SJ 92055 92,132)	Waste management facility: bailed separated materials	Yes
24	28 November 2013	Swindon Skips Limited, 1 Marshgate, Swindon, SN1 2PA (NGR: SU 16751 85,668)	Waste management facility	Yes
25	05 March 2014	Recovered Fuels Shipping Limited, Duncan Street Ind Estate, Duncan Street, Salford M5 3SQ (NGR: SJ 82168 97,953)	Waste management facility	Yes
26	21 July 2014	Aventes Recycling, Marshgate, Swindon. SN1 2PA (NGR: SU 16825 85,589)	Waste management facility	Yes
27	26 August 2014	Aventes Recycling, Marshgate, Swindon. SN1 2PA (NGR: SU 16825 85,589)	Waste management facility	Yes
28	14 November 2014	Bridge Foot Quarry Recycling Centre, Flint Cross, Royston, Hertfordshire, SG8 7PP (NGR: TL 40381 43,146)	Waste management facility	Yes
29	11 March 2015	Mitchelin's Farm, Rayleigh, Essex SS6 7NG. GR TQ 778908	Waste tire site.	Yes
30	8 October 2015	Wagstaffs Recycling Centre, Long Lane, Great Heck, Near Selby, North Yorkshire DN14 0BT. GR SE 59437 21,503	Waste management facility	Yes
31	02 March 2016	Global Hygiene, Drummond Road, Stafford, Staffordshire. GR SJ 9226 2487	Unregulated site.	Yes

^a Column 1, 'AQC', is used as the unique identifier throughout this paper.

^b National Grid References are approximate based on the description of the incident.

^c Site operation description is garnered from Environment Agency notes and local media reports.

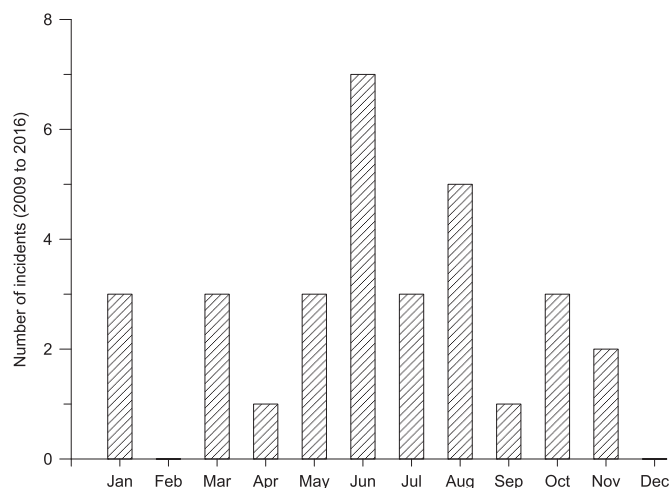


Fig. 1. Monthly distribution of AQCs over the period April 2009 to March 2016.

whisker plots (Grapher, Golden Software) and to generate rolling 24-h and 1-h averages that could be compared to existing short-term guideline values. The data was also used to provide a chronological analysis of the progression of individual fires.

3. Results and discussion

3.1. AQC incident details

Over the period April 2009 to March 2016, there were 31 major fire incidents across England and Wales that met the conditions to establish an AQC and be managed by the EA. For five of the incidents it was decided by the AQC that monitoring was not required, and for three incidents, data are missing, leaving PM monitoring data available for 23 major incident fires. A full list of the incidents, including date, location, and type of operation is provided in Table 4. For context, over the same period during which the 31 major fire incidents occurred, the Fire Service in England and Wales responded to 572,754 primary fires comprising: ‘dwellings’ (238,212), ‘other buildings’ i.e. commercial or industrial (127,697), ‘road vehicles’ (164,624) and ‘other outdoors’ (42,221) (Home Office, 2016) and so this highlights the rarity of fires of the scale and seriousness that would warrant an AQC to be established.

Regulated waste management operations are the most common function of sites where major incident fires occur, representing 74% of all AQCs over the period analysed. This category of site is likely to continue to be a concern for the foreseeable future, since EU/UK Government policy is for increased reliance on processing facilities to recover a range of materials such as tyres (e.g. AQCs 3, 4, 8, 17 and 29), plastics and waste electrical and electronic equipment (e.g. AQC 5). Guidance for the reduction of fire risk at waste management sites continues to develop in the UK, with the Waste Industry Safety and Health Forum updating its guidance in April 2017 (Waste Industry Safety and Health Forum, 2017). In addition, the EA has introduced a Fire Prevention Plan requirement at sites they regulate, further to European Union Directive 2010/75/EU on industrial emissions (Integrated Pollution Prevention and Control). Nevertheless, there are numerous ‘illegal’ sites that function outside of regulatory control entirely or do not adhere to conditions imposed by their regulator (e.g. AQC 31).

Of the incidents listed in Table 4, two posed additional concerns for public health: (1) AQC 9, which was a site recognized as requiring additional management due to the risk it posed from the chemicals stored on site under the European Seveso Treaty, and (2) AQC 10, because it handled waste labeled with a hazardous risk phrase, as defined under European Union Directive 2008/98/EC (note that monitoring data was available for AQC 9 but not AQC 10).

3.2. Seasonal distribution of AQC incidences

Evidence of a seasonal distribution of the AQCs is indicated by a higher incidence between June and August, as shown in Fig. 1. The cause of this is not discernible and may be that warmer weather conditions during these months contributes to the ignition and sustaining of the fires.

3.3. Particulate mass concentration monitoring

Figs. 2, 3 and 4 show box and whisker plots for the distribution of 15-min mean concentrations of PM_{10} , $PM_{2.5}$, and PM_1 , respectively, during each of the 23 major fire incidents for which monitoring data was available. Statistical outliers, indicated as points on the plots, are those values that exceed 1.5 times the upper quartile range; nevertheless, these values are included in the overall analysis because they are components of the measured data set and were used by the AQC in

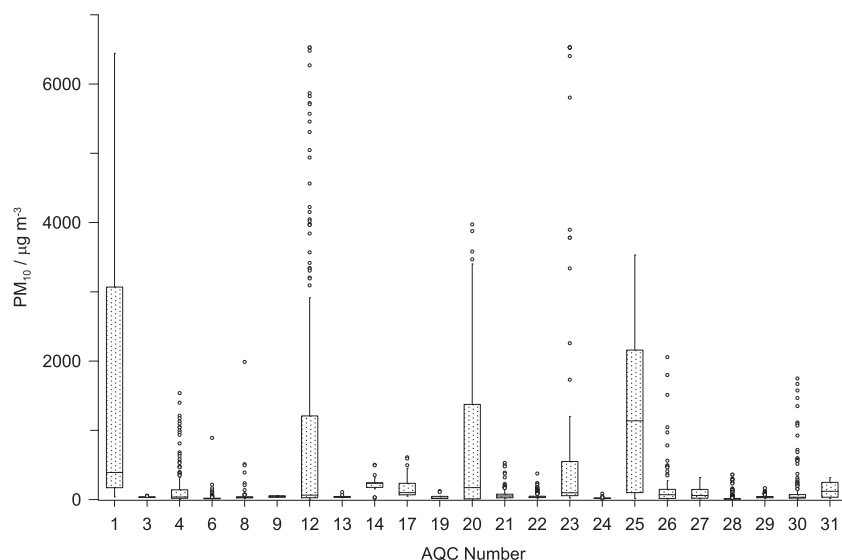


Fig. 2. Range and extent of PM_{10} concentrations as measured at AQCs. Shown are the range, including outliers, the 25th to 50th percentile range and the median point of all measurements.

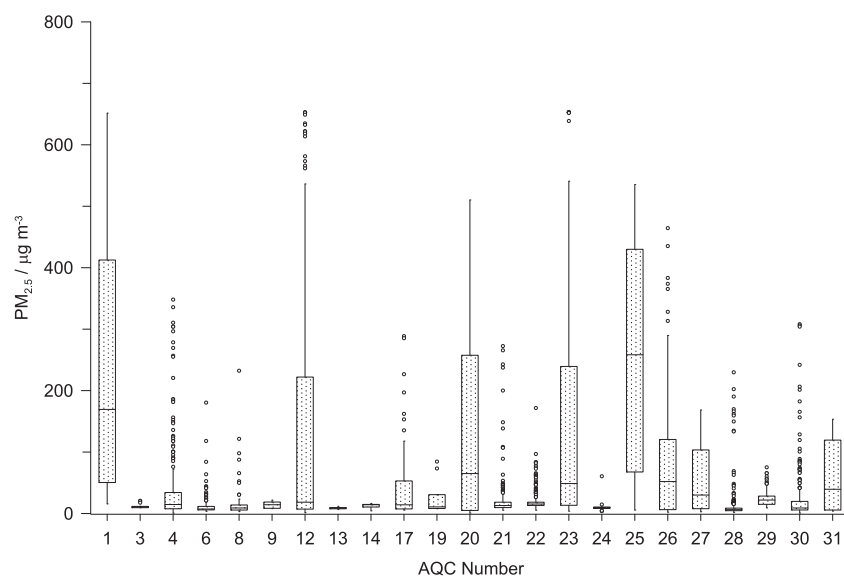


Fig. 3. Range and extent of $PM_{2.5}$ concentrations as measured at AQCs. Shown are the range, including outliers, the 25th to 50th percentile range and the median point of all measurements.

their public health risk assessment. Mean, median and maximum concentrations are summarised in Table 5. PM_{10} concentrations in excess of $6000 \mu g m^{-3}$ were observed in three of the incidents, with corresponding maximums of over $600 \mu g m^{-3}$ for $PM_{2.5}$ and PM_{10} . Whilst these are significantly elevated concentrations, it is important to note that (1) it is not possible to exclude the possibility that actual concentrations exceeded this, as the Osiris monitor has a concentration ceiling that is 'in excess of $6000 \mu g m^{-3}$ ', (Deary et al., 2016; Turnkey Instruments Limited, 2009), (2) the heated inlet may reduce the level of volatiles measured and potentially cause a shift in the distribution of size fractions and (3) atmospheric water vapour is noted as exaggerating PM_{10} values from the Osiris but not $PM_{2.5}$ and PM_{10} values. Mass concentration values of PM observed for the major incidents analysed, far exceeded those of most of the pollution episodes listed in Table 1 and emphasise the public health significance of these major air pollution incidents. Another important characteristic of these incidents, as illustrated in Fig. 5 for AQC 12, is the variability in PM emissions over time, largely due to the prevailing meteorology, as well as the

techniques applied to control the fire.

3.4. Applicability of Osiris particulate monitoring data for assessing potential short-term health effects

Since most epidemiological evidence and guideline values for PM correspond to periods of at least 24 h, the intermittent elevated short-term (15-min average) concentrations of PM observed during these incidents raises important questions about likely health impacts on exposed individuals and the corresponding public health response that is necessary for health protection. However, in order to adequately characterize the health risk associated with 15-min and 1-h average concentrations, we need to have confidence in the performance of the monitor over such timeframes, i.e. in its ability to generate responsive and accurate data compared to reference continuous methods, such as TEOM-FDMS.

The Osiris equipment is an 'indicative standard' (Waldén et al., 2010) for air quality monitoring as it does not utilise a reference

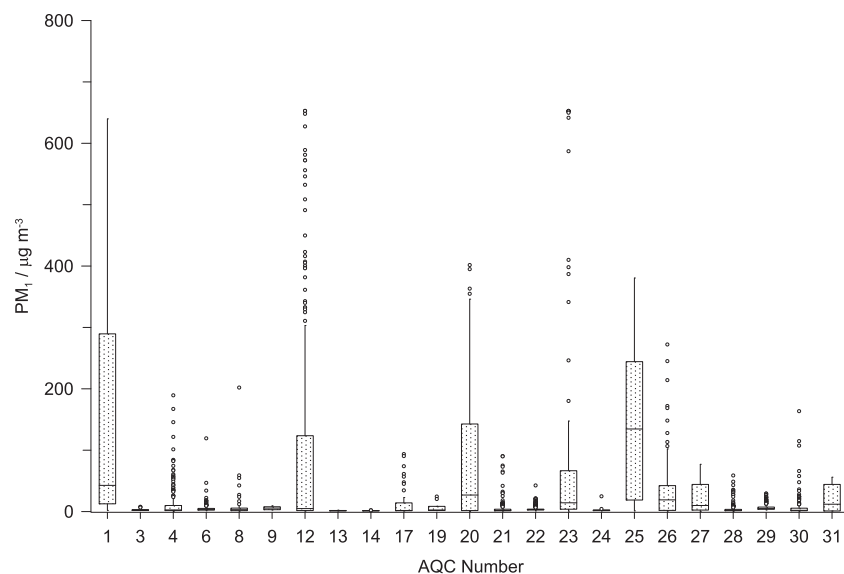


Fig. 4. Range and extent of PM_{10} concentrations as measured at AQCs. Shown are the range, including outliers, the 25th to 50th percentile range and the median point of all measurements.

Table 5Statistics for measured PM₁₀ and PM_{2.5} concentrations at AQC incidents over the period April 2009 to March 2016.

AQC No.	PM ₁₀ /μg m ⁻³			PM _{2.5} /μg m ⁻³			Number of 15-min periods in dataset
	Mean ± sd	Median	Max	Mean ± sd	Median	Max	
1	1450 ± 1933	392	6445	234.1 ± 208.80	169.4	651.5	42
3	37.59 ± 8.49	34.1	55.3	12.11 ± 3.94	10.71	20.75	15
4	142 ± 250.3	37.4	1537.2	38.71 ± 63.45	14.68	348.08	263
6	29.24 ± 75.36	14.75	890.93	13.10 ± 1.60	7.38	180.54	152
8	89.6 ± 258.5	32.1	1987.8	19.67 ± 34.98	8.97	232.42	65
9	45.03 ± 11.5	45.85	59.11	14.07 ± 5.15	14.25	21.77	7
12	921.5 ± 1564.5	64.7	6527.9	134.60 ± 185.90	18.5	652.8	255
13	47.76 ± 25.55	39.49	108.7	8.94 ± 1.66	9.11	11.76	9
14	229.8 ± 128.9	234.7	502.1	12.49 ± 3.96	14.25	16.46	19
17	179.4 ± 153.5	100.1	611	53.5 ± 79.9	14.2	288.7	39
19	40.6 ± 41.0	19.1	123.5	25.73 ± 28.92	10.83	84.88	10
20	752.1 ± 1036.0	172.5	3973.6	136.70 ± 148.30	65	510.2	152
21	68.39 ± 74.02	52.32	530.33	23.01 ± 39.02	12.97	272.45	222
22	51.27 ± 46.67	36.46	375.51	21.65 ± 19.67	15.42	171.68	207
23	1159 ± 2155	98	6528	163.9 ± 225.80	48.7	652.8	67
24	20.61 ± 9.65	19.57	86.76	9.94 ± 6.33	9.71	60.58	75
25	1204 ± 1095	1137	3530	243.10 ± 181.30	258.4	535.5	42
26	162.5 ± 316.7	69.8	2057.6	84.33 ± 102.98	52.04	464.33	120
27	79.21 ± 66.07	58.85	318.97	51.46 ± 48.39	30	168.42	128
28	24.76 ± 51.26	9.97	359.79	12.87 ± 28.48	6.26	229.85	353
29	40.37 ± 20.79	35.51	164.37	23.38 ± 10.86	21.89	75.28	326
30	118.5 ± 274	34	1748.5	26.03 ± 47.96	9.03	308.14	235
31	144.2 ± 113.2	119.9	316.4	60.4 ± 58.70	39.5	153.4	28

standard method for measurement of PM₁₀ and PM_{2.5} as detailed in BS EN 12341:2014 (noting this document does not provide a reference standard for the measurement of PM₁). ‘Indicative’ is a standard of equipment that is capable of confirming the presence of PM and will indicate a mass concentration value.

There have been several significant co-location studies carried out for the Osiris and the DustMate instruments, both of which use the same measurement system. A recent study by one of us compared DustMate and TEOM-FDMS, hourly-averaged PM₁₀, (range: 3.5 to 46.8 μg m⁻³) and PM_{2.5} (1.7 to 23.4 μg m⁻³) concentrations for 41 separate measurement periods (Deary et al., 2016). The slope (DustMate = slope x TEOM-FDMS) and intercept for PM₁₀ were 1.02 ± 0.06 and -3.7 ± 1.2, respectively ($R^2 = 0.73$) whilst for PM_{2.5}, the respective values were 0.78 ± 0.06 and -0.63 ± 0.55 ($R^2 = 0.79$). Over 15-min averaging periods, using an Osiris instrument, Gulliver and Briggs (2004) determined a slope of 1.03 ($R^2 = 0.83$) for PM₁₀ in a study that comprised 302 × 15-min intervals, equivalent to 75 h. In addition, unpublished data by us, comparing the DustMate with a TEOM instrument for PM₁₀ data averaged over 15-min periods (see Fig. S1 in Supplementary Material), gave a slope (DustMate = slope x TEOM) and intercept of 0.89 ± 0.04 and -1.82 ± 0.67 respectively ($R^2 = 0.71$, $n = 196$), though it should be noted that this was for a Nafion/desiccant drying system and not the heated inlet used in the other studies reported. The results of these studies and others reviewed in Deary et al. (2016) provide evidence that the Osiris/DustMate instrument is sufficiently responsive over 15-min and 1-h averaging periods. Moreover, for between-sampler comparability tests, conducted on two DustMate instruments, using one-minute averaged readings, a slope and intercept of 1.05 ± 0.03 and 0.36 ± 0.5, respectively ($R^2 = 0.73$) were obtained for PM₁₀, with corresponding values of 0.79 ± 0.01 and 0.19 ± 0.06 ($R^2 = 0.86$) for PM_{2.5}. This study demonstrates the responsiveness of Osiris/DustMate instruments to simultaneously measured environmental variations over very short sampling periods (Deary et al., 2016).

The underlying reason for the UK EA choosing the Osiris for use in the AQinMI service was a compromise, balancing the technical ability of the monitoring equipment against the practicability of deployment. In contrast, reference standard PM methods such as TEOM and TEOM/FDMS or the Grimm EDM180 (Approved for PM₁₀ and PM_{2.5} to

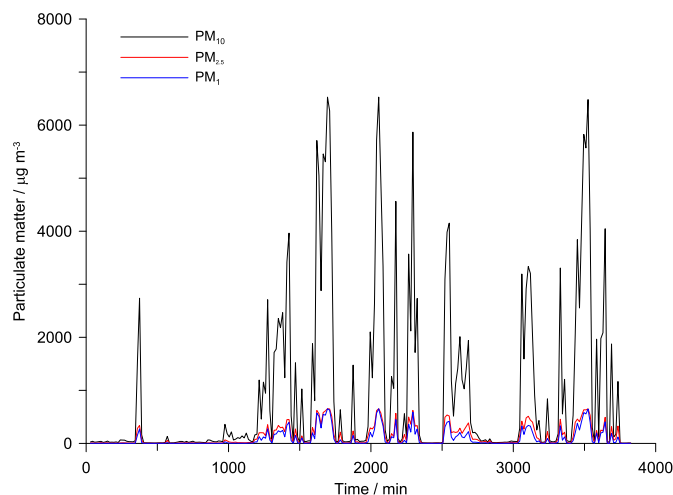


Fig. 5. Plot of measured PM₁₀, PM_{2.5} and PM₁ mass concentrations at AQC12. Values plotted represent 15-min mean measured values.

EN12341) require field deployment as part of a mobile lab. There are two such labs available to the AQinMI service, both using the Grimm EDM180, however, because of deployment and set-up time, they have had limited use during major incident fires. This aspect of the service is currently being re-evaluated, with trailer-based Grimm 180 instruments likely to be deployed more frequently in future.

3.5. Health implications of observed particulate concentrations

The main evidence of health effects arising from exposure to elevated PM concentrations over very short durations comes from a series of studies in which healthy volunteers were exposed to diesel fumes, or collected ambient PM, over 1 or 2-h periods. Subsequent bronchoalveolar lavages and biopsies (typically 6 h to 18 h after exposure), allowed the response in the upper and lower respiratory tract to be examined (Behndig et al., 2006; Ghio et al., 2000; Pourazar et al., 2005; Salvi et al., 1997; Salvi et al., 1999; Stenfors et al., 2004; Tong et al., 2014). Also of relevance, are studies on health effects reported

following smoke exposure in firefighters (Greven et al., 2012; Swiston et al., 2008) and on US soldiers exposed to smoke from burning oil wells during the first Gulf War (Smith et al., 2002). In addition, a limited amount of syndromic surveillance data exists for these elevated PM concentrations over short duration exposure incidents (Harper et al., 2009).

For volunteer studies, there is clear evidence of an induced inflammatory response in the upper respiratory tract (increased neutrophil levels) for short-term (1 to 2 h) PM₁₀ exposures ranging from 100 to 300 $\mu\text{g m}^{-3}$ (Behndig et al., 2006; Pourazar et al., 2005; Salvi et al., 1997; Salvi et al., 1999; Stenfors et al., 2004). For the same studies, the response in the alveoli appears to be confined to an increase in the proinflammatory proteins, IL-6 and IL-8 (Stenfors et al., 2004), or anti-inflammatory factors such as urate and glutathione (Behndig et al., 2006). Evidence has also been produced from these studies that the trigger for the release of the proinflammatory factors may be oxidative-stress (Kelly, 2003; Pourazar et al., 2005). For PM_{2.5}, a concentration-dependent alveolar (as well as bronchial) inflammatory response has been observed after a 2-h exposure to concentrations of up to 311 $\mu\text{g m}^{-3}$ (average upper quartile exposure of 206.7 $\mu\text{g m}^{-3}$) (Ghio et al., 2000). Volunteers in the same study showed elevated blood fibrinogen levels at 18 h; this is a factor that has been associated with ischemic heart disease.

The maximum 15-min PM_{2.5} concentrations encountered during major incident fires, analysed in Figs. 2 to 5, are of the same order of magnitude as the levels used in the volunteer studies on diesel exhaust fumes, and so we might expect similar immunoresponses. Surprisingly, when mild asthmatics were tested in volunteer studies (2-h exposure to a 108 $\mu\text{g m}^{-3}$ PM₁₀ diesel exhaust), there was no evidence of inflammation in either the upper or lower respiratory tract. This observation was ascribed to the increased production, compared to non-asthma sufferers, of IL-10, a downregulator of the proinflammatory factors IL-6 and IL-8 (Stenfors et al., 2004).

The literature on short-term acute respiratory effects in firefighters exposed to PM generally supports the findings from the volunteer studies, though the levels of exposure were an order of magnitude higher. The estimated exposure of firefighters to respirable PM (< 3.5 μm), using monitored CO measurements as a surrogate (Reinhardt and Ottmar, 2004), showed peak concentrations > 2200 $\mu\text{g m}^{-3}$ and 6 h of concentrations exceeding 1000 $\mu\text{g m}^{-3}$ (Swiston et al., 2008). Swiston et al. (2008) and Greven et al. (2012) found that whilst inflammatory responses, including increased levels of bronchial neutrophils, are observed, there is no significant effect on lung function, notwithstanding short-lived respiratory complaints that did not require medical intervention, such as coughs, nasal congestion and sputum production (Greven et al., 2012; Swiston et al., 2008). Greven et al. (2012) also found that proinflammatory factors such as IL-8 were elevated in the bloodstream for at least three months after exposure, though the authors could not rule out the influence of additional exposures between measurements. In a similar situation, soldiers exposed to acute episodes of elevated PM concentrations from burning oil wells in the 1991 Gulf War showed no significant increase in hospitalisation rates, though they represent a very healthy sub-group of the population, as do firefighters (Smith et al., 2002).

Syndromic surveillance data for the incidents covered in this paper are not available as far as we are aware. However in March 2009, before the AQinMI service commenced, syndromic surveillance data (calls to the UK National Health Service, regarding difficulty breathing, coughing and eye problems) was used during a major fire in London, whereby the reports of such symptoms were compared against a CHEMET prediction of plume direction. The data showed that there was no significant increase in reported symptoms in the area affected by the plume, despite estimates of concentrations in the hundreds of micrograms per cubic metre, or higher, at some locations (Harper et al., 2009).

Whilst the evidence from troop, firefighter and syndromic

surveillance studies suggest that exposure to significant concentrations of PM over short periods does not result in increased hospitalisation, at least in healthy subjects, the firefighter and volunteer studies do demonstrate clear evidence of an inflammatory response in the upper and lower respiratory tract, that may be long-lasting. In addition, there is epidemiological evidence from ambient studies showing that acute exposure to PM for 24-h periods or more has a robust and consistent association with mortality (0.5% increase per 10 $\mu\text{g m}^{-3}$) among the general population (World Health Organisation, 2006). From a consideration of all this evidence, a precautionary approach to PM exposure during major incident fires would seem appropriate.

3.6. Public health response

The PM data presented in Figs. 2 to 5 and summarised in Table 5 are considered indicative of the range of concentrations to which members of the public were exposed during such incidents. In addition to the technical concerns from the use of the Osiris monitor, it is important to acknowledge that there are a number of uncertainties and limitations with the data collection, including (1) the ability to monitor only two sites at any one time, with locations restricted to sites that have access to appropriate services; (2) monitoring contractors not being able to measure within the most concentrated area of the plume, for example inside any cordons established by the emergency services; and (3) non-uniform dispersal of PM within the plume, with the likelihood that some areas will be exposed to higher concentrations than others near the plume.

The default UK advice in major incident fires is to recommend sheltering indoors with windows and doors closed, where pollutant concentrations will be significantly reduced, and protection will be given against concentration peaks (Cabinet Office, 2013; Stewart-Evans et al., 2016). Most evacuations occur because buildings are already inundated with products of combustion such that it makes sheltering impossible, with that decision being taken by the emergency service commanders before the involvement of the AQO.

Stewart-Evans et al. (2016) discussed the application of health criteria to air pollutants released from fires, both for major incidents and extended burns. They note that for many chemical substances there are short-term emergency guideline values (10 min, 30 min, 1 h, 4 h and 8 h) that can be used, such as the Acute Exposure Guideline Levels (AEGs) of the US EPA (Environmental Protection Agency). However, for PM, there is currently no guidance corresponding to periods shorter than 24 h, although Lipsett et al. (2008) did derive 1 to 3-h guidelines based on the US Air Quality Index, as part of guidance on wildfire smoke developed for public health officials in the US. Nevertheless, the latest version of this guidance lists only the 24-h guidelines (The California Air Resources Board and California Department of Public Health, 2016).

In the absence of specific short-term guidance on exposure to fine PM, in Table 6 we have summarised 24 h guideline ranges, derived from various sources for different purposes, including: WHO guidelines (World Health Organisation, 2006), UK Air Quality Indices (AQIs) (Department of Environment Food and Rural Affairs, 2017), US AQIs (US Environmental Protection Agency, 2014), US wildfire smoke advice (Lipsett et al., 2008; The California Air Resources Board and California Department of Public Health, 2016) and UK advice in major incidents (Brunt and Russell, 2012). These give an indication of potential health impacts on healthy and 'at risk' members of the public, as well as providing advice on the appropriate public health action. Based on US advice during wildfires (Lipsett et al., 2008; The California Air Resources Board and California Department of Public Health, 2016), as well as UK advice derived from major incident response (Brunt and Russell, 2012), the consensus for closing schools and other public buildings ('Trigger to Close Public Buildings') is around 150–160 $\mu\text{g m}^{-3}$, whereas evacuation ('Trigger to Evacuate') should be considered at concentrations between 240 and 320 $\mu\text{g m}^{-3}$, though

Table 6Overview of the health effects, advice and recommended actions derived from various public health sources for 24-h exposure to PM_{2.5} and PM₁₀ at different levels.

PM ₁₀ /μg m ⁻³	PM _{2.5} /μg m ⁻³	Description	Health effects/Advice/recommended actions
50	25	UK/EU 24-h mean guideline for PM ₁₀ and WHO 24-h guideline for PM _{2.5}	Concentrations above the PM ₁₀ guideline correspond to UK Air Quality Index band for 'Moderate', which advises that "adults and children with lung problems, and adults with heart problems, who experience symptoms, should consider reducing strenuous physical activity, particularly outdoors."
75	37.5	WHO Interim target-3	Equates to an approximately 1.25% increase in short-term mortality over that for the short-term WHO Annual Quality Guideline (50 for PM ₁₀ μg m ⁻³ and 25 μg m ⁻³ or PM _{2.5})
76	54	UK AQI: High	UK AQI advice is that for concentrations exceeding these guidelines, "at risk individuals: adults and children with lung problems, and adults with heart problems, should reduce strenuous physical exertion, particularly outdoors, and particularly if they experience symptoms. People with asthma may find they need to use their reliever inhaler more often. Older people should also reduce physical exertion. General population: Anyone experiencing discomfort such as sore eyes, cough or sore throat should consider reducing activity, particularly outdoors."
100	50	WHO Interim target-2	Equates to an approximately 2.5% increase in short-term mortality over that for the short-term WHO Annual Quality Guideline (50 for PM ₁₀ μg m ⁻³ and 25 μg m ⁻³ or PM _{2.5})
101	71	UK AQI: Very High;	UK AQI advice is that for concentrations exceeding these guidelines, "at risk individuals: adults and children with lung problems, adults with heart problems, and older people, should avoid strenuous physical activity. People with asthma may find they need to use their reliever inhaler more often. General population: reduce physical exertion, particularly outdoors, especially if they experience symptoms such as cough or sore throat."
150	75	WHO Interim target-1	Equates to an approximately 5% increase in short-term mortality over that for the short-term WHO Annual Quality Guideline (50 for PM ₁₀ μg m ⁻³ and 25 μg m ⁻³ or PM _{2.5})
150.5	150.5	US AQI: Very Unhealthy;	US AQI advice is that for concentrations exceeding these guidelines, "schools: move all activities indoors or reschedule them to another day. Consider closing some or all schools. Cancel outdoor events involving activity (e.g., competitive sports). Consider cancelling outdoor events that do not involve activity (e.g. concerts)". If only PM ₁₀ concentrations available, then it can be assumed that PM ₁₀ is primarily comprised of PM _{2.5} and that the PM _{2.5} 24-h average should, be used.
160	–	UK Trigger to Close Public Buildings (Brunt and Russell, 2012)	Schools, nurseries, day-care centers and other similar facilities should be closed (to be interpreted and implemented in conjunction with other observations such as the vertical and lateral spread of the plume, whether it is grounding, etc).
250.5	250.5	US AQI: Hazardous	US AQI advice is that for concentrations exceeding these guidelines, "consider closing schools. Cancel outdoor events (e.g., concerts and competitive sports). Consider closing workplaces not essential to public health. If PM level is projected to remain high for a prolonged time, consider evacuation of at-risk populations". If only PM ₁₀ concentrations available, then it can be assumed that PM ₁₀ is primarily comprised of PM _{2.5} and that the PM _{2.5} 24-h average should, be used.
320	–	UK Trigger to Evacuate (Brunt and Russell, 2012)	When this 24-h average has been reached, and concentrations are predicted to continue at higher than this level for a further 24 h, then evacuation should be considered.

only if the elevated PM concentrations are expected to be at this level for 48 h or more (Stewart-Evans et al., 2016). As noted above, evacuations often take place before public health authorities become involved, with the emergency services electing to move people from their homes because of the proximity to the fire, or if properties are already contaminated with smoke, making sheltering pointless.

3.7. Using exceedances of 1-h exposure thresholds as predictors of longer-term exposure

24-h acute exposure thresholds exist for both PM₁₀ and PM_{2.5}, spanning a range of concentration-related guidance, as detailed in Table 6, and discussed in the previous section. If it could be shown that, during an incident, a breach of a 24-h threshold could be predicted from a corresponding breach in a 1-h threshold, this might provide a solution to the problem of having no reliable short-term guideline values for PM₁₀ and PM_{2.5}.

Figs. 6 and 7 show, for PM₁₀ and PM_{2.5} respectively, the results of an analysis whereby the predictive capability of 1-h thresholds ranging from 50 to 600 μg m⁻³, in 10 μg m⁻³ increments, is assessed against various 24-h thresholds of relevance to public exposure during major incident fires. For PM₁₀, perhaps the most important 24-h thresholds are those that correspond to closure of public buildings (160 μg m⁻³) and evacuation (320 μg m⁻³) (Brunt and Russell, 2012), though other guidelines from Table 6, ranging from 75 μg m⁻³ (WHO Interim

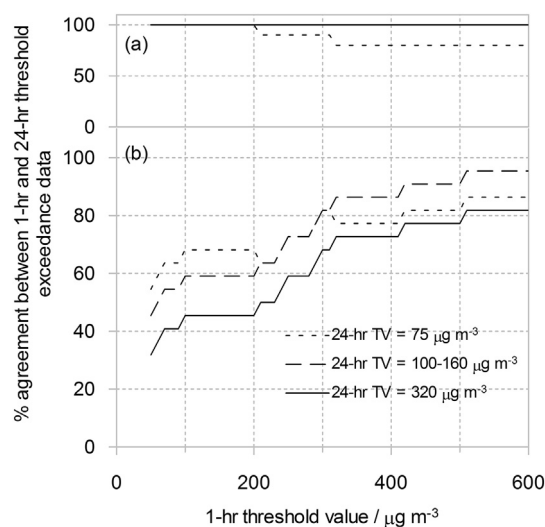


Fig. 6. Percentage agreement between within-incident exceedance of threshold values (TV) for rolling mean 1-h PM₁₀ concentrations and selected TVs of public health protection significance for rolling mean 24-h concentrations across 23 major incident fires. Panel (a) shows the percentage of incidents where exceedance of the 24-h TV was correctly predicted by 1-h TV. Panel (b) shows the percentage of incidents where the exceedance/non-exceedance of the 24-h TV was correctly predicted by 1-h TV.

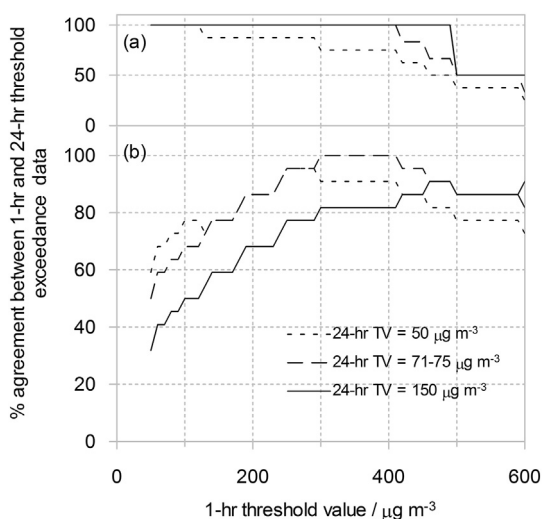


Fig. 7. Percentage agreement between within-incident exceedance of threshold values (TV) for rolling mean 1-h $\text{PM}_{2.5}$ concentrations and selected TVs of public health protection significance for rolling 24-h concentrations across 23 major incident fires. Panel (a) shows the percentage of incidents where exceedance of the 24-h TV was correctly predicted by 1-h TV. Panel (b) shows the percentage of incidents where the exceedance/non-exceedance of the 24-h TV was correctly predicted by 1-h TV.

Target-3) to $150 \mu\text{g m}^{-3}$ (WHO Interim Target-3) have also been included in the analysis. For $\text{PM}_{2.5}$, there are no separate closure or evacuation 24-h standards (PM_{10} concentrations can be used for this purpose). However, guidelines from Table 6, ranging from $50 \mu\text{g m}^{-3}$ (WHO Interim Target-2) to $150.5 \mu\text{g m}^{-3}$ (US AQI: Very Unhealthy) have been used in the analysis.

The predictive capability is based on two measures of correspondence between the rolling mean 1-h and 24-h thresholds for an incident, as detailed in panels (a) and (b) of Figs. 6 and 7. Panel (a) shows the percentage of incidents where an exceedance of a 1-h threshold correctly predicts a corresponding exceedance of a 24-h threshold. Panel (b) is the overall agreement between the two thresholds, i.e. the percentage of incidents where either (i) both thresholds are exceeded or (ii) both are not exceeded. In choosing the most advantageous 1-h threshold to use for public health protection purposes, we should maximise selectivity, i.e. the overall agreement shown in panel (b), whilst ensuring that all 24-h exceedances are correctly predicted. For both $\text{PM}_{2.5}$ and PM_{10} , the curves in panel (b) for all 24-h thresholds are similar in shape, i.e. a sharp increase in percentage agreement, followed by a plateauing-out at higher 1-h threshold concentrations, and in the case of $\text{PM}_{2.5}$, a subsequent decline in agreement, that corresponds to a reduction in the correct prediction of 24-h exceedances, as shown in panel (a).

For PM_{10} , choosing a 1-h threshold in excess of $510 \mu\text{g m}^{-3}$ will give probabilities of 95% and 82% respectively of predicting the correct 24-h outcome for 'closure' ($160 \mu\text{g m}^{-3}$, but including thresholds in the range 100 to $160 \mu\text{g m}^{-3}$) and 'evacuation' ($320 \mu\text{g m}^{-3}$) thresholds, whilst still ensuring a 100% prediction of the corresponding 24-h exceedances. It is noteworthy that Lipsett's, now withdrawn, 1 to 3-h average Recommended Action Level for the closure of public buildings and possible evacuation was set at $526 \mu\text{g m}^{-3}$ for $\text{PM}_{10}/\text{PM}_{2.5}$ (Lipsett et al., 2008). The 1-h predictive capability for the $75 \mu\text{g m}^{-3}$ threshold (WHO-Interim Target-3) is less reliable, principally because at 1-h thresholds above $200 \mu\text{g m}^{-3}$ the ability to predict the 24-h exceedances declines, as shown in Fig. 6(a). Nevertheless, a 1-h threshold concentration set at $150 \mu\text{g m}^{-3}$ for PM_{10} would give a 68% probability of predicting the correct 24-h outcome, whilst ensuring that all 24-h exceedances were predicted (Fig. 6(b)).

For $\text{PM}_{2.5}$, Fig. 7(b) shows that a 1-h threshold set at $350 \mu\text{g m}^{-3}$ will give probabilities of 100% and 82% respectively of predicting the

correct outcome of the $71 \mu\text{g m}^{-3}$ (and $75 \mu\text{g m}^{-3}$) and $150 \mu\text{g m}^{-3}$ 24-h thresholds, whilst still ensuring the ability to correctly predict all exceedances. As with the lower 24-threshold analysed for PM_{10} , the $50 \mu\text{g m}^{-3}$ 24-h $\text{PM}_{2.5}$ threshold is not reliably predicted because of the reduction in accuracy in the prediction of exceedances (Fig. 7(a)).

Whilst the analysis described in this section has yielded relationships that may have application in the public health response to major incident fires, the conclusions should be treated as tentative given the relatively small number of studies on which they are based. Nevertheless, the robustness of the analysis will be improved over time as more studies are included.

4. Conclusion

Major incident fires occur relatively infrequently, but they do have the potential to expose local populations to PM matter concentrations of up to thousands of micrograms per cubic metre. Whilst our analysis of 23 major incident fires shows that such extreme concentration excursions are of relatively short duration, there is evidence from volunteer studies, and from an analysis of respiratory effects in firefighters, that exposure to significantly elevated short-term concentrations is likely to result in an inflammatory response in the respiratory tract. Among the more vulnerable members of the population, this physiological response may exacerbate existing medical conditions, possibly necessitating hospitalisation, although we have found no evidence that this occurred in any of the 23 incidents analysed.

The UK's AQC process for monitoring and modelling emissions to the atmosphere from major incident fires is a positive development for characterizing the risk to public health posed by significant uncontrolled emissions to air and for that information to be disseminated to incident managers. What remains less developed are short-term exposure standards (minutes to hours) that the responders can use to determine the appropriate public health response, which includes decisions on evacuation and the need to provide pro-active health surveillance for those exposed to high levels of airborne pollution. Nonetheless, our analysis of the potential use of 1-h thresholds to predict exceedances of 24-h thresholds offers a way forward in the absence of very short-term guideline values. Our tentative 1-h thresholds for use in this context are $510 \mu\text{g m}^{-3}$ for PM_{10} and $350 \mu\text{g m}^{-3}$ for $\text{PM}_{2.5}$.

Our future work will further appraise the effectiveness of AQC monitoring and its role in public health risk assessment. There is a clear need to (1) analyse monitoring data for gaseous emissions during the 23 AQC incidents; (2) appraise the AQC monitoring data alongside reference standard monitoring where available, such as from fixed local authority and national air quality stations, should they coincide; (3) combine sources of monitored fire emission plumes from fixed air quality stations using standard monitoring equipment (i.e. TEOM instruments) and spot levels from Fire and Rescue Service teams, where it is available, to provide a clearer understanding of modelled emission plume levels at ground level; (4) to model the levels measured from the various sources and back-calculate to source to identify levels within the plume itself, (5) to consider the correlation between levels of measured pollutant and health data using the UK's syndromic surveillance system and (6) to appraise further the relationship between short-term high level exposure to thresholds and longer term standards.

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